

Synthesis and Characterization of New Complexes of (N-P-Amino Diphenyl Amine) Amino (2-Hydroxy Phenyl) Acetonitrile Ligand with Some Transition Metal Ions

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Abstract: The synthesis and characterization of new complexes of Fe(II), Co(II), Ni(II) and Cu(II) with bidentate (N-p-amino diphenyl amine) amino (2-hydroxy phenyl) acetonitrile ligand has been described. The ligand was prepared from 2-hydroxy benzaldehyde and 4-aminodiphenyl amine in the presence of KCN and acidic medium. The complexes were synthesized by treating an ethanolic solution of the ligand with appropriate amount of metal salts [1:2] [M: L] ratio. The complexes were characterized by using metal and elemental analysis, infrared and electronic spectra, molar conductivity, magnetic susceptibility, and mole ratio method. According to the obtained data the probable coordination geometries of Iron, Cobalt, Nickel and Copper in these complexes are octahedral. All complexes were found to be non-electrolyte in absolute ethanol, and the complexes were formulated as $[ML_2Cl_2]$.

Keywords - α -aminonitrile, Cobalt, Copper, Complexes, Iron and Nickel.

I. Introduction

Strecker reaction [1], is the oldest known synthesis of α -aminonitriles since 1850 when Adolph Strecker interacted an aldehyde, an amine and hydrogen cyanide. α -aminonitriles synthesis represents one of the simplest and most economical methods for the preparation of α -amino acids [2,3] for both laboratory and industrial scales [4]. The synthesis of Nitrogen-containing heterocyclic is important such as thienopyrimidine derivatives which have antibacterial and antifungal activities higher than the corresponding antibacterial ampicillin and antifungal nystatin [5-7]. On the other hand some of the α -aminonitrile derivatives like 1-amino-4-phenylnaphthalene-2-carbonitrile have been postulated to have high fungistatic activity even stronger than the activity of the commercial fungicide – Kaptan [8].

Moreover, among many other applications, they are readily hydrolyzed to diamines, and diamine compounds are of interest as ligands for Platinum (II) complexes with potential antitumor properties [9,10].

Complexes with ligands N_2 -type are of interest not only in theoretical but also from a practical point of view. They have found application as antitumor activity [11], antibacterial activity [12] and antiviral activity in agriculture field [13].

To prepare α -aminonitriles generally an imine is reacted with a cyanide source. Notable among them are HCN [14], KCN [15], $(EtO)_2P(O)CN$ [16,17], Et_2AlCN [18,19], Bu_3SnCN [20,21], and Me_3SiCN [22]. Recently, several modifications of the Strecker reaction using catalysts have been reported which includes mesoporous aluminosilicate (Al-MCM-41) [23], lanthanum(III)-binaphthyl disulfonate [24], nanocrystalline magnesium oxide [25], BINOL-phosphoric acid [26,27], $Fe(Cp)_2PF_6$ catalyst [28], Jacobsen's thiourea catalyst [29], *N*-heterocyclic carbene (NHC)-amidate palladium(II) complex [30], $Yb(OTf)_3$ -pybox [31], K_2PdCl_4 catalyst [32], gallium (III) triflate [33], bisformamides [34], superparamagnetic iron oxide [35], Trimethylsilyl Cyanide – Iodine catalyst [36], Trimethylsilyl Cyanide-*p*-Toluene sulphonic acid [37], homogeneous and solid – supported promoter [38] and $K_5CoW_{12}O_{40} \cdot 3H_2O$ as heterogeneous catalyst [39]. Finally, an efficient and practical method for the Strecker reaction under eco-friendly conditions have been achieved to processed organic reactions in water or ionic liquid in the view of green methodologies [40-43]. In this work α -aminonitriles have often been used as chelating ligands through two nitrogen atoms in the field of coordination chemistry. This is one of the first studies to reveal a new coordination field of α -aminonitrile compounds as ligands and their interactions with transition metal ions.

II. Experimental

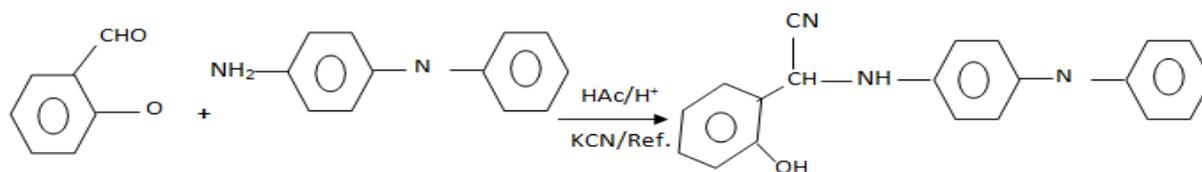
All the chemicals and solvents used for the synthesis were of reagent grade and were obtained commercially from Fluka company with the exception of $FeCl_2 \cdot 4H_2O$, $CoCl_2 \cdot 6H_2O$, $NiCl_2 \cdot 6H_2O$ and $CuCl_2 \cdot 2H_2O$ salts were obtained from British Drug House (BDH). The infrared spectra of the ligand and the complexes were recorded on a Shimadzu (8300) FTIR Spectrophotometer, as CsI disk (ACROS organics COMPANY). Electronic absorption spectra were recorded in the range (200 – 1100) nm on a Shimadzu (UV-1650 Pc) UV-Vis spectrophotometer in freshly prepared 10^{-3} M in absolute ethanol at room temperature using

quartz cell (1.00) cm. Atomic absorption technique was used to determine the metal contents of the complexes using a (GBC 933 plus) Atomic absorption spectrophotometer. Molar conductivity was used to measure the conductivity of the complexes at room temperature in freshly prepared 10^{-3} M in absolute ethanol using (WTW inoLab) Digital conductivity meter. Elemental analysis for Carbon, Hydrogen and Nitrogen elements were carried out using EuroEA Elemental Analyzer. Gallen Kamp melting point apparatus was used to determine the melting points of the ligand and the prepared complexes.

2.1 Preparation method of (N-p-amino diphenyl amine) amino (2-hydroxy phenyl) acetonitrile ligand [L]:

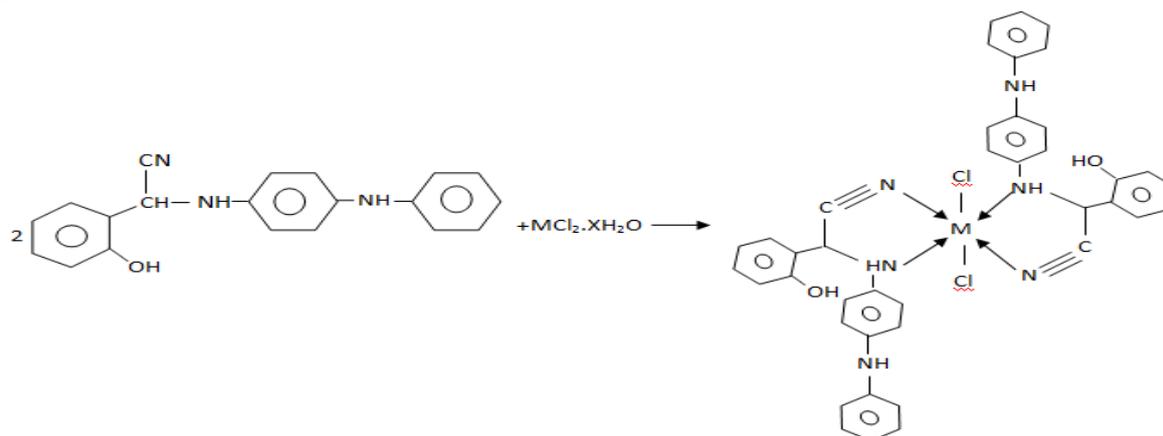
The method that adopted for [L] preparation can be described as follows: The 2-hydroxy benzaldehyde (1 mmol) was added to (20 ml) glacial acetic acid, *p*-toluene sulphonic acid was added in very small portion as catalyst, followed by addition of (1 mmol) *p*-amino diphenyl amine. The pH was adjusted to about 4 by addition of concentrated sulfuric acid drop wise to obtain Schiff base which stirred for 15 min. Potassium cyanide (2 mmol) was added to the mixture and kept under stirring for 4 days. The reaction mixture was poured into ice and then made slightly alkaline with ammonium hydroxide solution. The desired solid precipitate which formed was filtered, washed with water and air dried.

The presence of nitrile group in the prepared α -aminonitrile was indicated by treating few amount of the sample with 10% sodium hydroxide solution, the liberation of ammonia after hydrolysis of nitrile group was detected by wet red litmus paper. Purity of the obtained compound was checked by TLC, using chloroform and ethyl acetate (1:1) as eluent. The product color was dark green and the yield percentage was (88 %) and the melting point was (78d) $^{\circ}$ C. The reaction was clarified below:



2.2 Preparation method of metal complexes:

The complexes [ML₂Cl₂] have been prepared by the reaction of 2 mmol (0.630 gm) of L with 1mmol of metal chloride (0.199 gm, 0.238 gm, 0.238 gm and 0.171 gm for FeCl₂.4H₂O, CoCl₂.6H₂O, NiCl₂.6H₂O and CuCl₂.2H₂O respectively) dissolved in (20 ml) absolute ethanol and refluxed with stirring under anhydrous conditions for 4 hours. The obtained complexes were collected after evaporation of ethanol and triturated with petroleum ether (60-80 $^{\circ}$ C) then filtered. The products were left in the desiccators to be dried under P₂O₅. The general reaction was clarified below:



III. Results And Discussion

All the compounds reported in this work are presented in table (1) which illustrated physical properties along with their molar conductivity at room temperature. Metal and elemental analyses of all compounds are represented in table (2). The ligand (L) was characterized by elemental analysis and infrared spectral data. α -aminonitrile compounds are bifunctional like amino acids but with nitrile group instead of carboxylic, (L) compound considered trifunctional due to the presence of hydroxyl group in benzene ring which could be analogue of serine amino acid. Therefore the expected coordination type of (L) is tridentate which resembles the behavior of some amino acid like histidine [44], but the obtained complexes were characterized as bidentate(N₂- type). The complexes have been prepared under anhydrous conditions to avoid any hydrolysis of

nitrile group to amide due to the presence of water molecules and metal ion [45].when organic molecules are coordinated to metal cations in organo-metallic or coordination compounds, they become susceptible to nucleophilic attack like water molecules which cause hydrolysis of nitrile into amide and this could be avoided under anhydrous condition [46]. In spite of using anhydrous conditions complexation of zinc metal ion with (L) was not successful and dragged into hydrolysis of nitrile group to amid which clearly characterized in FTIR chart at 1656cm^{-1} . Therefore the complex dose not mentioned. These prepared complexes were stable in the solid state and were characterized by the usual methods; IR and UV- Visible spectroscopy, Molar conductivity, metal and elemental analysis, magnetic susceptibility and mole ratio.

On the basis of the characterization methods it was suggested that all complexes probably have octahedral structure with coordination number 6 that includes two atoms of coordinated chlorides and without coordinated or lattice water. The conductivity values for the complexes of (10^{-3}M) in absolute ethanol were recorded in rang (11-16) indicating that the non-electrolytic nature of the complexes [47]. Mole ratio method was used to determine the ratio of metal ion to ligand for the complexes at λ_{max} ($\lambda_{\text{max}} = 881\text{nm}$ for $[\text{FeL}_2\text{Cl}_2]$, $\lambda_{\text{max}} = 566\text{ nm}$ for $[\text{CoL}_2\text{Cl}_2]$ and $\lambda_{\text{max}} = 680\text{nm}$ for $[\text{CuL}_2\text{Cl}_2]$) in alcoholic solutions.

3.1 Infrared spectroscopy:

The infrared spectral data of [L] in table(3) showed some characteristic bands related to α -aminonitriles ; Like $2240 - 2260\text{ cm}^{-1}$ which could be attributed to $\text{C}\equiv\text{N}$ group[48]. A red shift of the range $35-95\text{ cm}^{-1}$ was observed for $\text{C}\equiv\text{N}$ stretching vibration on coordination due to the decrease of bond order as result of metal nitrogen bond formation. A red shift of N-H group on complexes could not be detected due to the presence of O-H group which usually over shade it. On the other hand the spectrum of the complexes showed new bands around $451-488\text{ cm}^{-1}$ and $376-393\text{ cm}^{-1}$ due to $\nu\text{ M-N}$ and $\nu\text{ M-Cl}$ respectively [49].

3.2 Ultra violet – visible spectroscopy:

The data of ultra violate spectra of the ligand and the complexes and the suggested structures are listed in table(4).The ligand showed two absorption bands in the region 39840 cm^{-1} and 32051 cm^{-1} corresponding to ($\pi \rightarrow \pi^*$) and ($n \rightarrow \pi^*$) transitions respectively. Shifting has been observed in ligand field after complexation and new bands were observed due to the presence of metal ions in the complexes.

The electronic spectrum of Iron (II) complex showed one broad band at 11350 cm^{-1} due to (d-d) transitions. A band at 26666 cm^{-1} in the spectrum represented the (L \rightarrow M) charge transfer for the $[\text{FeL}_2\text{Cl}_2]$ complex [50]. The effective magnetic moment was (5.19 BM) which is typical for distorted octahedral [51]. The electronic spectrum of Nickel (II) complex showed three (d-d) transition bands at 22421cm^{-1} , 20080 cm^{-1} and 10493 cm^{-1} corresponding to ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{1g}(\text{P}) (\nu_3)$, ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{1g}(\text{F}) (\nu_2)$ and ${}^3\text{A}_{2g}(\text{F}) \rightarrow {}^3\text{T}_{2g}(\text{F}) (\nu_1)$ transitions respectively. Which suggested mostly octahedral geometry.The magnetic moment value for the Nickel (II) complex was found to be (2.90 B.M) [51, 52]. A peak of $[\text{NiL}_2\text{Cl}_2]$ complex spectrum appeared at 25575 cm^{-1} which represented to the (L \rightarrow M) charge transition band[50].Cobalt(II) complex has magnetic moments value (3.78 B.M) which agreed with the expected value for the high spin Cobalt (II) ion in octahedral environment[51,53].

The electronic spectrum of Cobalt (II) complex showed two main bands at 17667 cm^{-1} and 16611 cm^{-1} . These two bands are assigned to ${}^4\text{T}_{1g}(\text{F}) \rightarrow {}^4\text{T}_{1g}(\text{P})(\nu_3)$ and ${}^4\text{T}_{1g}(\text{F}) \rightarrow {}^4\text{A}_{2g}(\text{F})(\nu_2)$ transitions respectively. The (ν_1) transition is expected to appear at large wave length. Thus it could not be observed, therefore it was calculated. The observed transitions are consistent with an octahedral geometry. A band of $[\text{CoL}_2\text{Cl}_2]$ complex spectrum appeared at 25445cm^{-1} represented to the (L \rightarrow M) charge transition band [50].The electronic spectrum of Copper (II) complex showed two bands. These two bands appeared at 14705 cm^{-1} and 19230 cm^{-1} ascribed to the (d-d) transitions. Another band appeared above 26666 cm^{-1} represented to the (L \rightarrow M) charge transition [50]. The obtained value of the effective magnetic moment (1.82BM) is typical for distorted octahedral Cu(II) chelates [51,53].

Table (1) Physical properties of (L) and its metal complexes with their conductivity values

Compound	Color	M.P. °C	Yield %	$\Omega^{-1}\text{cm}^2\text{mol}^{-1}$
L	Dark green	78d.	95.23	-----
$[\text{FeL}_2\text{Cl}_2]$	brown green	121-123	79.36	16
$[\text{CoL}_2\text{Cl}_2]$	Brown	99-102	81.57	12
$[\text{NiL}_2\text{Cl}_2]$	Light green	223-225	84.25	14
$[\text{CuL}_2\text{Cl}_2]$	Dark blue	117-120	83.77	11

Table (2) Analytical data of (L) and its Complexes

Formula M.Wt. (g/mol)	Elemental And Metal Analysis Calculated (Found)			
	C%	H%	N%	M%
	L 315	76.19 (76.03)	5.39 (5.38)	13.33 (13.21)
[FeL ₂ Cl ₂] 756.8	63.42 (60.93)	4.49 (4.22)	11.10 (10.85)	7.37 (7.00)
[CoL ₂ Cl ₂] 759.9	63.16 (62.86)	4.47 (4.31)	11.05 (10.77)	7.75 (7.44)
[NiL ₂ Cl ₂] 759.7	63.18 (62.96)	4.47 (4.22)	11.05 (10.72)	7.72 (7.53)
[CuL ₂ Cl ₂] 764.5	62.78 (62.66)	4.45 (4.41)	10.98 (10.77)	8.31 (8.00)

Table (3) The characteristic bands of FTIR spectra of the ligand and its metal complexes in (cm⁻¹)

Compound	ν O-H cm ⁻¹ ν N-H cm ⁻¹	ν C-H cm ⁻¹ Aromatic	ν C-H cm ⁻¹ Aliphatic	ν C≡N Nitrile	Aromaticity ν	δ C-H out of plane deformation		ν M-N	ν M-Cl
						ortho	Para		
L	3356	3053	2930	2210	1595,1510	750	827	-----	-----
[FeL ₂ Cl ₂]	3380	3056	2923	2115	1595, 1504	750	831	451	380
[CoL ₂ Cl ₂]	3334	3075	2936	2172	1595,1506	752	829	488	376
[NiL ₂ Cl ₂]	3396 v. bro	Obscured	2960	2175	1614,1512	752	880	474	393
[CuL ₂ Cl ₂]	3400	3078	2940	2165	1578,1512	754	850	480	382

Table (4) Electronic spectra in absolute ethanol for the (L) and its metal complexes.

Comp.	λ nm	ν cm ⁻¹	Transition	B ⁰ cm ⁻¹	B ⁻ cm ⁻¹	β	Dq/ B ⁻	10Dq cm ⁻¹	15B ⁻ cm ⁻¹	Suggested structure
L	251 312	39840 32051	$\pi \rightarrow \pi^*$ $n \rightarrow \pi^*$							
[FeL ₂ Cl ₂]	233 301 375 881	36232 33223 26666 11350	Ligand Field Ligand Field C.T $^5T_{2g} \rightarrow ^5E_g$							distorted O.h
[CoL ₂ Cl ₂]	211 289 393 566 bro. 602sh. 1268	47393 34602 25445 17667 16611 7881(cal)	Ligand Field Ligand Field C.T $^4T_{1g}(F) \rightarrow ^4T_{1g}(P)$ $^4T_{1g}(F) \rightarrow ^4A_{2g}(F)$ $^4T_{1g}(F) \rightarrow ^4T_{2g}(F)$	971	709.2	0.73	1.23	8730	10638	O.h
[NiL ₂ Cl ₂]	263 293 391 446 498 mean 953	38022 34129 25575 22421 20080 10493	Ligand Field Ligand Field C.T $^3A_{2g}(F) \rightarrow ^3T_{1g}(P)$ $^3A_{2g}(F) \rightarrow ^3T_{1g}(F)$ $^3A_{2g}(F) \rightarrow ^3T_{2g}(F)$	1030	734.8	0.71	1.43	10493	11022	O.h
[CuL ₂ Cl ₂]	247 295 375 521 sh. 680 bro.	39370 33898 26666 19230 14705	Ligand Field Ligand Field C.T $^2B_{1g} \rightarrow ^2B_{2g}$ $^2B_{1g} \rightarrow ^2E_g$							distorted O.h

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